Progress in Microchannel Steam Reformation of Hydrocarbon Fuels

2003 Hydrogen and Fuel Cells Merit Review

May 19-21, 2003

Greg A. Whyatt

Team Members: Kriston Brooks, Jim Davis, Chris Fischer, Dave King, Larry Pederson, Susie Stenkamp, Ward Tegrotenhuis, Bob Wegeng

Pacific Northwest National Laboratory



Approach –2003

- A STATE OF THE STA Confirm benefits of operating steam reformer at higher temperature.
 - Productivity for benchmark fuel increased 3X between 650°C and 850°C
 - Sulfur tolerance significantly increased.
 - Higher productivity reduces thermal mass → less air and fuel required for startup, reduced losses associated with cycling thermal mass on startup.
 - Operate a low dP steam reformer capable of fast startup
 - Design/build system to achieve fast start while maintaining capability to achieve 78% efficiency during steady-state operation.
 - Fast start to be achieved by maintaining pressure drops low and using large flow of combustion gas. Initial work based on a 650°C reformer in stainless steel.
 - Integrate reformer with WGS and PROX at 2 kWe scale.



Objectives

And the state of t Develop a prototype microchannel-steam reforming based fuel processor at 2 kWe scale that will meet DOE performance targets when scaled up to 50 kWe. 2003 Projected 2005 DOF

	2000 1 10,0000	2000 000
Performance Criteria	Performance	Target
Power Density	1191 W/liter	700
Specific Power	376 W/kg	700
Efficiency	78.7%	78.0%
Durability	>1000 hr ^(a)	4000 hr
Transient (10% to 90%)	~5 sec ^(a)	5 sec
Cold Start, 20C	30 sec ^(a)	< 1min

⁽a) Durability and transient demonstrated for reformer and associated HX only. Cold start estimate is for reformer and HX only.

- Engage industrial partner(s) to facilitate development of full-scale fuel processing system.
- Develop reactors, vaporizers, recuperative heat exchangers, and condensers broadly applicable to other fuel processing options.





FY 1998
Full-size gasoline vaporizer/combustor
R&D100 Award

Project Timeline



FY 1999
Fast SR kinetics
observed in a
microchannel reactor

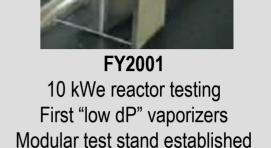


FY 2000

Designed and built

10 kWe SR with integrated

HX network





Engineered catalyst, reactor development

Feasibility of rapid start capability

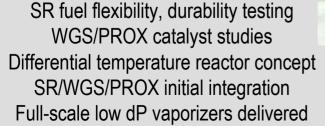
Sulfur management



Investigate improved steam reforming kinetics and sulfur tolerance at high temperatures. Fast start. WGS and PROX integration.

Integrated reformer/fuel cell at ~2 kWe

Collaborate with industrial partner(s) on manufacturing, field testing, lifetime, controls



Battelle

FY 2002

2003 2004

2005

2006

2008

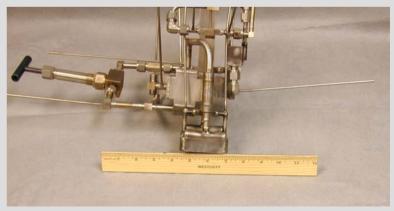
Pacific Northwest National Laboratory
U.S. Department of Energy

High Temperature Steam Reforming Motivation for Investigation

- ► Reduce Reactor Size exponential productivity increase with temperature
- ► Reduce Catalyst Quantity higher productivity results in less catalyst
- ► Reduce Reactor Startup Mass reduction in mass results in less fuel and air compression required during startup. Losses associated with cycling of thermal mass are reduced.
- ► Improve Sulfur Tolerance Improve sulfur tolerance may allow sulfur cleanup to be performed on reformate rather than on liquid fuel.
- ► Evaluate Carbon Deposition Potential High temperatures increase potential to deposit carbon by cracking of hydrocarbons at inlet (even if not an equilibrium condition).

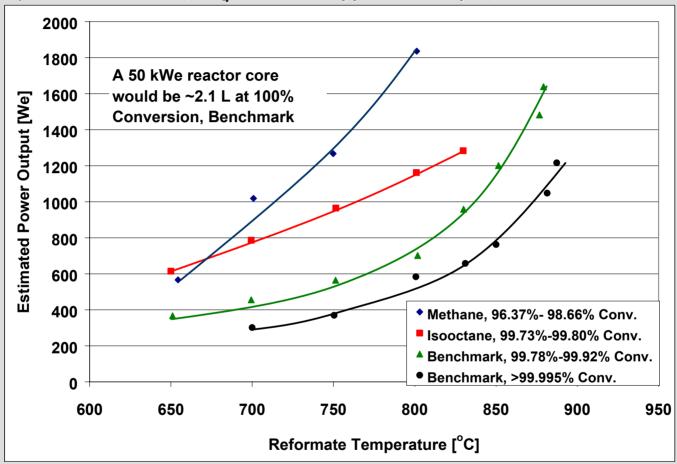
Inconel 625 Reformer







The State of the S Reformer Productivity vs Temperature (All fuels at 3:1 S:C, CH₄ conv. is % approach to equil., Inconel 625 reformer)



Stability of catalyst productivity verified by measuring the conversion at a reference condition before and after generation of the data for each curve.



6/11/2003

Characterization of Sulfur Sensitivity

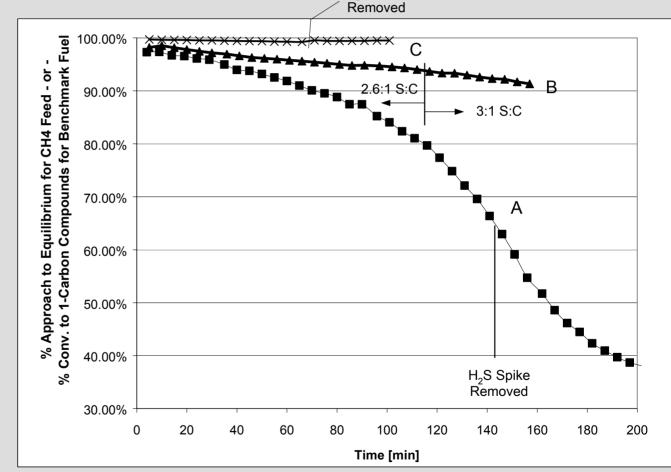
Sulfur Added as H₂S Spike in Methane

(3:1 S:C, Inconel 625 reformer)

A — Reformate outlet at 650°C. CH₄ with H₂S at 5.4 ppmv (10 ppmw benchmark equivalent)

B — Reformate outlet 650°C. Benchmark Fuel w/estimated 10 ppmw S plus CH₄/H₂S spike at 10 ppmw S equivalent.

C — Reformate outlet 800°C. Benchmark Fuel w/estimated 10 ppmw S plus CH₄/H₂S spike at 10 ppmw S equivalent. Sulfur from previous test (B) not removed prior to test.



Benchmark Fuel Composition, wt%: 74% Isooctane, 20% Xylenes, 5% methylcyclohexane, 1% 1-Pentene



Fast Start Approach

- ► Focus initially on steam reformer, vaporizer, and air and reformate recuperators. Incorporate WGS and PROX in system as development continues.
- Current system designed to 2 kWe at 650°C reformer temp.
- High combustion gas flows at low pressure drop heat the reformer, vaporizer and air recuperator.
- ▶ Reformate recuperator is heated via steam from vaporizer.
- ► Combustion gas at startup generated by non-catalytic atomized fuel combustion. Combustor tested for isooctane and regular gasoline. Provides 800°C gas in as little as 2 seconds.



Pacific Northwest National Laboratory U.S. Department of Energy

Concept for Low Pressure Drop, Rapid-Start Steam Reformer - Air Side Flows

And the second s Reactor Panels Vaporizer Panel Normal Blower Startup Combustor Start-up Blower Startup Air Recuperator Exhaust Reactor Panels Vaporizer Panel Normal Blower Start-up Blower Air Recuperator Exhaust Operation Fuel Introduction Points **Combustion Catalyst**



6/11/2003

2 kWe Reactor System – Predicted Air Side Pressure Drops

	2 kWe, Steady State Output Condition		30-Second Startup Condition	
Device	Flow [slpm]	∆P [inches H ₂ O]	Flow [slpm]	∆P [inches H ₂ O]
Steam Reformer Panel	100	0.65/panel 2.6 total for 4	800	5.2
Water Vaporizer Panel	100	0.28	800	2.2
Air Recuperator	100	1.5 cold side 1.6 hot side	200 (vap exhaust)	3.0
Total	100	6.0	4000	5.2

50kWe-Scale Air Compression Power Implications:

During operation theoretical compressor power is very low (~63W)

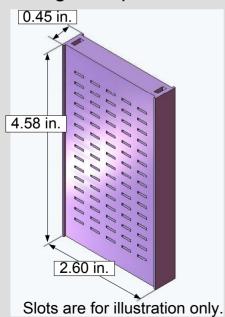
During 30 second startup, theoretical compressor power is ~2.2 kW



Fast Start/Low dP Reforming System Components

Reformer

- ♦4X, ~500 We panels for 2 kWe total at 650°C.
- lacktriangle Air side $\Delta P \sim 0.65$ inches water at 500 We output.
- uniform temperature for all reaction pathways.
- ◆Stainless construction, ~1534 g for 4 panels.

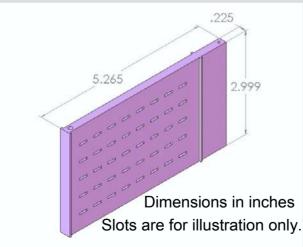


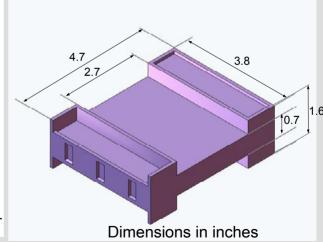
Water Vaporizer

- Low gas side ∆P, <0.28 in. water</p>
- Utilizes reactor exhaust
- ► Gas inlet/outlet 633°C/146°C
- Steam outlet 293°C
- ► ~400g

Air Recuperator

- ▶ Low ∆P, <2 in. water per pass</p>
- ▶ 87% effective, ~50°C outlet
- Excellent Turndown
- **≻** ~876 g







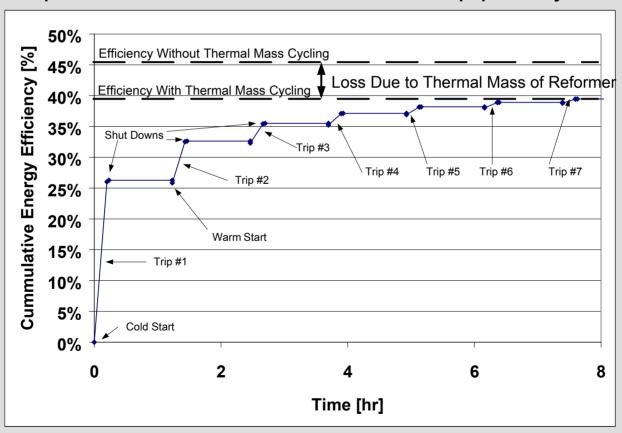
Preliminary Estimate of Efficiency Loss Due to Cycling Thermal Mass of Reformer on Startup/Shutdown Cycles

- ➤ Objective Assure that the thermal mass associated with the fuel reformer does not impose a severe efficiency penalty when thermally cycled in an automotive application.
- California Air Resources Board (CARB) real-world study
 - 6.7 Trips per day
 - 61% of cold soaks <1 hour
 - 24.8 mph (average per trip)
 - 46 miles per day
- Various Assumptions →7 trips/day separated by 1 hour shutdowns, each trip having 12 minutes at 37% output and 1.9 minutes at 5% output.
- Estimated masses of components with 3.5 inches conventional insulation, neglect connections.
- Calculation addresses only thermal mass effect, does not include electrical consumption and exhaust heat losses during startup.



Thermal Mass Startup Penalty

- Efficiency penalty ~5.2% for one day, ~4.5% for 7 day cycle.
- ► At 650°C reformer temperature, reactor is over half the thermal mass.
- Higher temperature reformer will reduce startup penalty





6/11/2003

Plans 2003

- And the state of t Sulfur Testing - Continue high-temperature sulfur testing to include longer-term testing, with liquid S spikes up to ~30 ppmw.
 - Improved Catalyst Improved catalyst is now available. It is expected to have similar activity at lower cost with increased mechanical and high-temperature durability.
 - Fast Start Low-dP System
 - Complete fabrication of stainless reactors and components
 - Conduct initial testing of panel reformer with new catalyst.
 - Generate design input for high-temperature low-dP reformer.
 - Conduct 30-second start tests. Initial tests will not include WGS and PROX.
 - Move to High-Temperature System Revise fast start system design to incorporate high temperature low-dP reformer.
 - Integrate with WGS and PROX at 2 kWe scale.



Interactions -

Microchannel Recuperator and Mixer Assembly for FASTER Project

Microchannel Recuperator

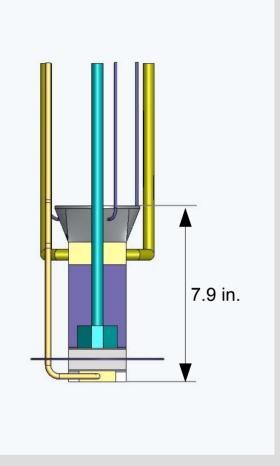
- -100°C hot end approach
- -3.6 kW duty

Carlo and the second se

- -Approx 1.1 kg w/o mixer
- $-\Delta P_{cold} + \Delta P_{hot} < 0.4 \text{ psi total}$
- -Uniform outlet temperature

Microchannel Mixer

- -Uniform outlet temp
- -Uniform air-reformate mixture





Interactions, continued

- ▶ Posting on the Federal Business Opportunity Website for Microchannel Fuel Reformation for Fuel Cell Applications. – The quality and quantity of responses led to a decision to license intellectual property on a non-exclusive basis.
- Testing of a microchannel water vaporizer designed for a 50 kWe fuel processor was concluded at McDermott. Similar testing concluded at GTI for boiler application.
- ► Finalizing contract with the Gas Technology Institute (GTI) to work with Cleaver-Brooks and GTI to provide a prototype micro-channel economizer for a highly efficient boiler.



Conclusions

- ▶ High Temperature Reforming Benefits
 - >3X productivity when increasing from 650°C to >850°C.
 - Significantly increased sulfur tolerance at 800°C vs 650°C.
 - Reductions in reformer thermal mass
 - Less air and fuel required during startup
 - Penalty associated with cycling thermal mass reduced
- ► A low pressure drop steam reformer that can start in 30 seconds appears feasible.
- Penalty associated with thermal mass of fuel processor is non-negligible. Penalty is reduced significantly by considering multiple restarts in a single day.



Responses to Comments

- "Given the ...thermal masses, startup energies need to be ...considered, not just start-up times": Calculation performed to assess impact of thermal mass on system efficiency.
- "Steam reformer needs to operate at higher temperature thus transition from stainless steel higher-temperature alloys needs to be completed": High-temperature Inconel 625 reactor tested up to 887°C.
- "Hydrocarbon conversion levels for liquid fuels are higher than earlier 90% but significant additional improvement is needed" -and-"High temperature reformer operation and demonstration of >99.9% conversion of non-methane hydrocarbons are critical to further progress": Reformer was operated on Benchmark Fuel over a range of temperatures to generate a curve of productivity possible while producing no detectable non-methane hydrocarbons.

